Natural-Matrix Radionuclide Standard Reference Materials

1 Purpose

This procedure describes the production, measurement and reporting procedures for the massic activity of radionuclides in natural-matrix Standard Reference Materials (SRMs).

The natural-matrix SRMs provide the metrology community with means by which they can: 1) validate their radioanalytical methods, 2) control the quality of their measurement process, 3) compare measurement results within projects, programs, within laboratory, and between laboratories over an extended period of time, and 4) support the traceability and credibility of measurement results [1].

2 Definitions

FWHM:	Full Width at Half Maximum
ICRM:	International Committee on Radionuclide Metrology

3 Scope

This procedure includes the selection of material matrices, processing them into final form, the interlaboratory comparison used to develop the data file from which the certified radionuclide massic activity will be derived, the data evaluation process, the NIST radiochemical analysis process to contribute to the data file, and the writing of the certificate.

4 Safety

Radiation safety: Rooms marked by Health Physics with magenta and yellow strip tape have been designated as Radiation Areas. Specific requirements for entry and exit from the rooms are provided by the NIST Health Physics Office.

Chemical safety: Chemical safety and training operations are provided by the NIST Safety Office.

5 Procedures

5.1 Quality Control

5.1.1 Document Change

Laboratory books shall be amended according to the policies set forth in Guide IRD-G-06.

5.1.2 Subcontracting

Reference materials certifications are not subcontracted (see NIST-QM-I Section 4.4.4).

5.1.3 Purchasing Services and Supplies

Purchased supplies will be inspected and their identity confirmed prior to use. The quality of the laboratory reagents will be confirmed during the analysis of reagent blanks to check the titer of the reagents, as well as any significant contribution to the blank signal. Any failure of the reagent's titer or indication of blank contamination will result in the suspension of use of the material, and a suitable replacement will be obtained to carry out the requested measurements.

Failures of a reagent's titer or blank contamination shall be noted in the investigator's laboratory notebook. Resolution of the failure will also be noted in the investigator's laboratory notebook.

Purchasing documents shall include technical specifications for all quality-critical labware, reagents and instrumentation. Quality-critical purchased items will be inspected and evaluated against the purchasing technical specifications

Quality-critical consumables of suppliers will be evaluated against technical specifications. Those suppliers which can supply consumables of adequate quality will be listed for the NIST purchasing agent to contact for bids.

5.1.4 Records

Analytical procedures, and instrument and software laboratory books and manuals shall be readily available at workstations as resources. When not being used, these resources shall be stored in nearby drawers that have been designated for such use.

Analytical records used for a project shall be sufficiently detailed to repeat analyses, investigate discrepancies, troubleshoot methodologies, and should include:

A. investigator's name
B. appropriate identification;
C. scope;
D. description of the type of item to be tested or calibrated;
E. parameters or quantities and ranges to be determined;
F. equipment, including technical performance requirements;
G. reference standards and reference materials required;
H. environmental conditions required and any stabilization period needed;
I. description of the procedure, including:
    a. affixing of identification marks, handling, transporting, storing and preparation of items,
    b. checks to be made before the work is started,

c. checks that the equipment is working properly and, where required, calibration and adjustment of the equipment before each use,
d. the method of recording the observations and results, and
e. any safety measures to be observed;

J. criteria and/or requirements for approval/rejection;
K. data to be recorded and method of analysis and presentation; and
L. the uncertainty or the procedure for estimating uncertainty.

5.1.5 Technical Requirements

5.1.5.1 Accommodation and environmental conditions

Environmental conditions are not critical factors that affect the quality of analyses. Laboratory activities will cease when the electricity goes out, a water pipe bursts, or temperature varies by more than human tolerance (i.e., $< 15^{o}$C or $> 32^{o}$C).

Tests and calibrations shall be stopped when the environmental conditions jeopardize the safety of the investigators.

Work is segregated among specified laboratories based on low (µBq/g - mBq/g), intermediate ($\geq$ Bq/g) and high levels ($\geq$ kBq/g) of sample radioactivity to prevent cross-contamination. Swipes of intermediate and high level laboratories are taken to confirm the absence of contamination at the conclusion of the sample preparation stage, and results are posted for all users of common area laboratories to determine the acceptability of the laboratory for use. Investigators entering intermediate or high level laboratories are not allowed to work in the low-level laboratories for at least 2 days after leaving the higher level area.

5.1.5.2 Methods

Radioanalytical methods used will be a blend of radiochemical techniques to provide measurement samples of adequate purity for unambiguous beta- and alpha-particle measurement. The appropriate selection of radiochemical techniques shall be from among standard methodologies and techniques described in evaluated literature citations. In cases where no standard methods are deemed appropriate, in-house methods are developed, verified against SRMs, CRMs, interlaboratory comparisons or proficiency testing results, and proficiency of use by each person involved in the project verified to fit the need and purpose of the measurement. These in-house methods are described in project files that should include:

A. investigator's name
B. appropriate identification;
C. scope;
D. description of the type of item to be tested or calibrated;

E. parameters or quantities and ranges to be determined;
F. equipment, including technical performance requirements;
G. reference standards and reference materials required;
H. environmental conditions required and any stabilization period needed;
I. description of the procedure, including:
    a. affixing of identification marks, handling, transporting, storing and preparation of items,
    b. checks to be made before the work is started,
    c. checks that the equipment is working properly and, where required, calibration and adjustment of the equipment before each use,
    d. the method of recording the observations and results, and
    e. any safety measures to be observed;
J. criteria and/or requirements for approval/rejection;
K. data to be recorded and method of analysis and presentation; and
L. the uncertainty or the procedure for estimating uncertainty.

5.1.5.3 Estimation of uncertainty of measurement

The procedure for combining and estimating uncertainties is defined in NIST Technical Note 1297 (1994). See also section 5.10

Electronic data collection, data transfers, appropriate use of use of software, and contributions by impurities are evaluated during spectral evaluations, quality control checks and statistical evaluations of the results. Any suspect results are investigated for root cause for adjustment, re-measurement, or classified as a marked error.

Measurement results are stored in file cabinets that are secured from intrusion by key-access locked room doors (when unattended), badge-only access locked building doors, and the NIST physical security system.

5.1.5.4 Equipment

All radiation measurement instruments in Building 245, Rooms B001, C10, C13, C15, and C17 have met purchasing technical specifications, are sufficiently shielded so that background radiation does not make any significant impact on quality of the measurements, and are monitored for stability of background, energy calibrations and efficiency calibrations to assure that they do not impact the quality of the measurements. The instruments are safeguarded from unauthorized adjustments by locking the rooms when unattended, badge-only access locked building doors, and the NIST physical security system.

Alpha-particle detectors are calibrated for energy response using historical data, and are confirmed using single or multi-nuclide electrodeposited sources before use. High-purity germanium gamma-ray detectors are energy calibrated using multi-gamma-ray line

sources, and are calibrated for efficiency using gravimetrically spiked matrix sources (see also section 5.5.8.1.2). In general, it is necessary that only the gain for the sodium iodide detectors be adjusted using gamma-ray emitting sources of known energy so that all energy lines of interest from samples are included for relative wide-open energy window analysis. Gas-flow beta-particle detectors are efficiency calibrated using gravimetrically prepared traceable sources as described in section 5.5.8.2.1. Background and quality control samples are measured for each specific project. All results are recorded and entered in the project folder.

All users of the instrumentation and associated software are expert with their operations. Instruction manuals are kept by the Radioactivity Group SRM custodian or available near the instrument. A backup copy of all commercial computational software that is provided on electronic media is used as the laboratory working copy. The original commercial software is stored in a metal cabinet in Building 245, Room C10.

A laboratory book shall be kept with each set of instruments that include information on:

A) manufacturer's name, type identification and NIST property number;
B) history of background and check source measurement;
C) location;
D) history of calibrations, adjustments, critical parameter settings; and
E) history of damage, malfunction, modification or repair.

5.1.5.5 Measurement traceability

All radiation measurement instruments are calibrated with NIST SRM traceable sources, gravimetrically prepared, that match the counting geometry, z_{eff}, self-absorption and scattering of measurement samples (see section 5.8.1.2 and 5.8.2.1).

Criteria for acceptable verification of traceability of derived sources is 1% u_c, and percent difference limited to 3%.

Alpha- and beta-particle emitting calibration and check reference sources are stored in pill boxes (245/C13) and planchet holders (245/B001) for safe storage.

5.1.5.6 Sampling

From one to three sub-samples of the natural-matrix materials are taken from each of five sample bottles for measurement; within bottle and between bottle heterogeneity is assessed when three sub-samples are taken from each of the five bottles of material. The sample size is selected to provide sufficient number of counts (few thousand to few tens of thousands of counts per radionuclide) for a reliable measurement in a reasonable amount of time (1-45 days). For natural-matrix materials, three sub-samples from each of five bottles to assess within and between bottle heterogeneity in addition to the massic activity for each radionuclide. Drying protocols are defined and performed when absorbed moisture significantly affects the determination of the samples massic activity

and uncertainty. Since sample dissolution and radiochemical purification steps are time and labor intensive, sequential separations are desired to also assess inter-radioisotope systematics as an additional check for measurement consistency and accuracy.

Statistical tools used to assess measurements of solutions and spiked samples include mean, standard deviation, and normal probability plots. Additional statistical tools used to assess natural-matrix radionuclide reference materials are discussed in section 5.5.7

5.1.5.7 Handling of test and calibration items

This is not applicable to this procedure.

5.1.5.8 Assuring the quality of test and calibration results

Measurements of test samples are planned, the plans are reviewed and discussed among cognizant colleagues for input and suggestions that include sample handling, sub-sampling, standard tracers, traceability, blank and background controls, sample dissolution options, radiochemical cleanup options, counting source preparation, measurement optimization, data analysis and reporting of results. The dissolution, cleanup, and measurement technique options are balanced to optimized the measurement uncertainty against time and cost restrictions that are acceptable to the client.

Control over the measurement process include analysis of blanks, comparison against reference values, interlaboratory comparisons, decay curve and spectral analyses for impurities, spectral resolution, inter-nuclide systematics, and statistical identification and investigation of suspect results and method bias.

5.1.5.9 Reporting the results

The format for reporting results is given in section 5.5.11, and examples of an SRM approval form and certificate are provided in Appendix C and D, respectively.

Statements of compliance to ANSI N42.22 and ANSI N13.30 acceptance criteria are used for environmental and bioassay radioanalyses as agreed upon by the customer.

5.2 ICRM'77 Recommendations

The International Committee on Radionuclide Metrology is composed of NMI and Secondary calibration laboratory representatives with considerable metrology experience. In 1977, the Low-Level Working Group of the ICRM convened to address the needs for low-level environmental radionuclide reference materials. Recommendations from this workshop included developing long-term (> 10 years) supplies of SRMs that preserve the natural characteristics of the material for the recommended samples (Bq/kg) as listed in Appendix A (Hutchinson and Mann). Natural-matrix SRMs that have been certified are listed in Appendix B.

5.3 Process Control

The responsibilities of the Radioactivity Group Leader, SRM Coordinator and SRM principal investigator as well as the coordination of SRM activities among them, is described in IRD-QM-II Sections 4.2.3.2.2 and 4.2.3.2. The natural-matrix SRM principal investigator relies on the SRM Coordinator to provide non-technical support services for the SRM such as development and production funding, labeling and storage of the SRM units, pricing, and advertising and marketing.

Throughout the following SRM matrix preparation procedures, each step is carefully considered and planned to maintain a reasonable degree of control. It is recognized that the preparation procedure will result in material alteration, however, it is desirable that the material characteristics and radionuclide speciation are preserved as much as possible. Care is taken to minimize contamination of the material by foreign sources (equipment wear, equipment cleanliness), and undue heating (e.g., flash evaporation instead of controlled freeze drying). As much as possible, a NIST representative accompanies the material if it is sent for commercial processing to inspect the equipment before use for cleanliness and after processing for equipment wear, and to obtain temperature charts and particle size information to assure that the material is processed as expected. By careful process control of the preparation of the material, it is expected that there will be minimal affects on the final certified values which are mainly derived from the measurement process of the packaged material.

5.4 Matrix Selection

Periodically, focused workshops are held with the radionuclide metrology sub-community to reconfirm the original ICRM'77 recommendations for target matrices, and to obtain recommendations for applicable radionuclide massic activities and suggestions for potential collection areas for the materials. For example, the 1995 workshop on SRMs for ocean studies focused on standards that would be crucial for establishing international comparability and credibility of data collected from monitoring around all nuclear-waste dump sites in the world's oceans. Priority actions include: 1) development of biota (shellfish, fish flesh, and seaweed), sediments (high calcium, high organic, high clay), and seawater media for ^{90}Sr, ^{137}Cs, ^{210}Pb, $^{239+240}$Pu and other radionuclides at concentrations of 0.03 to 10 Bq/kg; 2) certification of the reference materials through laboratory comparisons by a network of highly qualified international reference laboratories; and 3) coordination of standards production with the International Atomic Energy Agency programs. The low-level environmental radioactivity SRM issued, Ocean Sediment (SRM 4357), will be followed by additional SRMs of shellfish and seaweed. Up to three thousand bottles of each SRM are produced to provide the radiochemistry community with an anticipated 10 year supply of reference material.

5.5 Matrix Collection

Matrix collection procedures are consistent with 5.7.2 of NIST QM-I. Once the radionuclide composition and massic activity characteristics of the target matrix and potential collection sites are identified in 6.2, cost estimates are obtained from personal contacts with access to potential collection sites. At times, several tons of wet material may need to be collected, shipped and processed so as to eventually end up with the anticipated 10 year supply of the reference material. These factors are weighed to select the "best" material for the amount of "seed" money allotted for the development of the material into a SRM.

Purchase Orders are sent to the collection contacts to collect and send the material to NIST for processing and, when necessary, to field screen and dry the material prior to shipment.

5.6 Matrix Preparation

5.6.1 Size of SRM Unit

The minimum SRM unit size is chosen to be at least three times the typical sample size used by the metrology community for an analysis of the matrix. Typically, the maximum sample size for soils and sediments is 10 g; for biological materials, it is the amount of material that would result in 10 g of ash.

5.6.2 Drying

When economically feasible, the preferred method of initially drying the matrix is by lyophilization. The lyophilization procedure is monitored to assure that the material is kept below -20 ^{o}C throughout the process. In cases where lyophilization is unnecessary, air drying is acceptable.

5.6.3 Matrix Size Reduction

Initial size reduction of the matrix is necessary to remove large objects (e.g., rocks) that do not easily fit into the inlet for the pulverization equipment used in the next step. Size reduction may be a simple gross sieving of the material, and/or using a course mill (e.g., swing arm blade mill, or ball mill), followed by appropriately sieving the milled material. Care must be taken to minimize contamination of the matrix by excessive wear of the mill by selecting carbide or tungsten wear-resistant cage and blade materials.

It is during this initial matrix size reduction step when, if two sources of materials are to be combined to yield the desired SRM characteristics, the materials are initially mixed. The Human Lung, Human Liver, Ashed Bone and Ocean Sediment SRMs were composites of materials from two or more sources.

5.6.4 Pulverization

To minimize heterogeneity and particle size separation, pulverization of the material to µm size using narrow particle size dispersion is necessary. Air-jet pulverization is the method of choice for dry, friable materials. The average particle size using the air-jet pulverizer is 10 µm. The greatest source of material loss occurs during this operation because of the inefficiency of removing the finely powered material from the large volume of air used during the pulverization, and the losses need to be considered during the matrix collection so as to assure sufficient material for the anticipated 10 year supply of the SRM. Even this technology does not provide sufficient particle size reduction if radioactively "hot" particles are much smaller, e.g., 1 µm, as was encountered with the Rocky Flats Soil - I (SRM 4353).

The Human Liver and Lung materials were cryogenically hammer milled because of the relatively high oil content of the original material (although use of a cryogenic shatterbox mill would have reduced the particle size of the final product, it would not have been practical to mill the hundreds of kilograms of the original material). Nevertheless, micrometer-sized "hot" particles survived cryogenic hammer-milling as was evident in the Human Lung material (SRM 4352).

5.6.5 Blending

To additionally minimize heterogeneity, materials are thoroughly mixed, particularly to evenly disburse "hot" particles. In all cases, the blender of choice is the "V"-cone blender with a high-speed internal pin-type intensifier bar. One hour of blending has been adequate to achieve a good blend, as evidenced by heterogeneity evaluation measurements throughout the course of the twenty-five year history of certifying the natural-matrix radionuclide 4350 series SRMs.

Ideally, the material is blended as one batch. In cases where the capacity of the blender is insufficient to blend the entire amount of material, sub-batches are blended. In this case, additional blending of the split sub-batches is necessary. It is recommended that a three-level cross blending approach provides sufficient mixing of the final material.

5.6.6 Final Drying

Biologically-derived materials, such as the Human Lung, Liver and Seaweed, require final drying to remove moisture absorbed during material processing and to increase shelf-life. Lyophilization using industrial food drying protocols is the method of choice prior to final packaging (see also 5.6.2).

5.6.7 Packaging

Environmental materials are packaged in 250-mL polyethylene bottles, and biological materials are packaged in amber glass bottles (under dry nitrogen). The Human Lung and Liver materials are additionally sealed under vacuum, and stored in deep freezers (generally at about -70 ºC) until shipped to customers.

5.6.8 Sterilization

Sterilization of the final SRM product is necessary to satisfy export requirements of environmental and biological materials, and to increase shelf life. The materials are irradiated with ^{60}Co gamma rays to an absorbed dose in excess of 40 kGy. This absorbed dose is confirmed by dosimetry traceable to NIST provided by the vendor. The excessive sterilization provided by the 40 kGy absorbed dose precludes the need for microbiological analysis. The container materials are selected to avoid the deleterious effect of radiation damage to the bottles and caps.

5.7 Interlaboratory Comparison

Leading experienced international radiochemical metrology laboratories are invited to participate in a "best effort" replicate analysis interlaboratory comparison to obtain the most accurate and consistent radionuclide concentration data possible. The laboratories are selected through their excellence in scientific publications in the open literature, presentations at conferences, proficiency tests, interlaboratory comparisons, and scientific trustworthiness established through lengthy personal interactions and collaborations with Radioactivity Group staff members. All laboratories agree to participate without anonymity. Each laboratory is sent five SRM units for analysis and ^{229}Th, ^{232}U, ^{242}Pu and ^{243}Am chemical yield monitors. Each laboratory selects the radionuclides for which results are returned to NIST, and uses the radioanalytical methodology for which they are most expert. One measurement from each of the five SRM units is requested for each reported radionuclide. The laboratory also has the option of reporting three measurement results from each of the five SRM units for assessment of between bottle and within bottle heterogeneity. The laboratories are requested to report their results within one year of receiving the SRM units.

The information requested for reported analytical results include: five massic activity results for each radionuclide with the associated standard combined uncertainties, the reference date, and a brief description of the analytical method used.

5.1.7 Measurement Traceability

Each participating laboratory establishes traceability to the SI through appropriate (i.e., geometry, matrix, single-point radionuclide calibrations or calibration curve, and uncertainty analysis) calibration of instruments (e.g., high-purity germanium spectrometers) and gravimetrically diluted tracers with appropriate SRMs and CRMs from reputable NMIs.

5.8 NIST Analysis

5.8.1 Gamma Ray Measurements

5.8.1.1 NaI Measurements

The heterogeneity of the environmental material is initially evaluated by gross gamma-ray measurements using a 12.7 cm diameter NaI detector in 245/C15. The detection system is set for at least a 50 - 1350 keV energy window. Five to ten samples of 100 - 300 g are placed in glass or plastic (plastic is preferred if Compton background reduction is needed to optimize measurement sensitivity) counting containers made of flat-plate bottoms and cylinder walls cut from a single length of tubing, tamped to the same height, then sealed in the container. If electrostatic charge is not a problem keeping the material at the bottom of the counting container, Plexiglas is the preferred material for the container to minimize the ^{40}K contribution to the gamma-ray Compton background. A waiting time of 3 to 4 weeks is allowed for the sample to equilibrate in the container before measurement.

Each sample is then placed in a counting jig to assure that each sample is counted in the same geometry with respect to the detector. Each sample is counted for the same amount of live time, generally one day to one week per sample.

Check sources and blank sample containers are generally counted before, between samples and after the last sample is counted to ascertain the stability of the counting system during the measurement schedule.

5.8.1.2 HPGe Calibration

Gamma-ray measurements using HPGe provide nuclide-specific detection. HPGe systems in 245/C10/C13/C17 are used for quantifying the gamma-ray emitting radionuclides. Providing there is sufficient radioactivity, the same samples used for the heterogeneity assessment in 5.8.1.1 can be used for massic activity quantification.

Each sample is placed in a counting jig to assure that each sample is counted in the same geometry with respect to the detector. Each sample is counted for the same amount of live time, generally 2 to 4 weeks per sample.

Check sources and blank sample containers are counted before, between samples and after the last sample is counted to ascertain the stability of the counting system during the measurement schedule.

The HPGe instrument is calibrated for energy response and efficiency with a sample of the matrix of interest that has been quantitatively spiked with traceable SRM radionuclides of interest and thoroughly blended [2-4]. The thoroughness of blending is verified by measuring a set of 5 - 15 g sub-samples from the blended material. This method of calibrating the detector avoids the added uncertainty contributions that affect interpolation of efficiencies from an experimentally determined calibration curve.

5.8.1.3 Gamma-ray Measurement QC

NaI and HPGe instrument stability are monitored with check source and blank determinations during the scheduled sample measurements. The check sources are counted in a reproducible geometry and are used to check that the energy responses of the detectors have been maintained under control. The absolute value of the emission rate need not be traceable so long as they are predictable. The instrument blanks are empty counting bottles that are measured in the same geometry as the samples. These determinations are done at the beginning of the schedule, between samples and after the final sample is counted.

5.8.1.4 Data Analysis

Background corrected NaI spectra counts are manually summed over broad peaks, generally over the 50 - 2000 keV region of interest.

HPGe spectra are analyzed for net peak area above the Compton background using numeric methods that incorporate the assessment of peak shape and peak overlap [5-6]. Each time software is changed, it is verified by comparing computational results against previously validated results, and the change is recorded in the instrument's log book, which is accessible to all staff. The instrument log books note the current software version being used or acceptable for use.

5.8.1.5 Computations

The NaI net region of interest count rates for each peak from the replicate samples are compared using a Normal Probability Plot to evaluate the normality of the distribution of the data. When the probability plot correlation coefficient is greater than the statistical critical value, it is judged that the data distribution is consistent with a normal distribution and that the energy window's heterogeneity can be estimated using its relative standard deviation.

Massic activity determinations from the HPGe spectra are computed from the measurement model that includes contributions from the net peak counts, counting time, blank correction, mass of the sample, calibration based on the spiked standard sample, emission probability, and radioactive decay from the reference date.

5.8.2 Alpha- and Beta-particle Emitting Radionuclide Measurements

5.8.2.1 Isotope Dilution

Measurement of alpha-particle emitting radionuclides requires radiochemical separation and purification from the host matrix prior to measurement. The generally complicated chemical purification steps used result in varying degrees of loss of the target radionuclides. Isotope dilution with a known amount of an isotope of extremely low

abundance in the sample matrix provides a means for monitoring the loss of the analyte during the chemical processing steps.

The fundamental requirement is that the added traceable SRM isotope be chemically and physically equilibrated with the analyte radionuclide prior to any losses. This requirement is generally satisfied by total dissolution of the sample in an appropriate solvent (usually an acid solution) containing the tracing isotope (also called "tracer"). Further steps to ensure chemical equilibrium of analyte and tracer after dissolution include the use of strong oxidizing or reducing agents.

Losses of pure beta-particle emitting radionuclides, such as ^{90}Sr, can be traced using isotopes such as ^{85}Sr. Alternatively, addition of a known amount of the non-radioactive element of interest can be used as a "chemical yield monitor". Stable (non-radioactive) strontium is used to trace the ^{90}Sr losses during chemical purification steps. The amount of stable strontium recovered at the end of the radiochemical procedure is gravimetrically compared to the known amount originally added at the beginning of the procedure to determine the chemical yield. A correction to the amount of recovered stable element at the end of the chemical procedure is needed when the sample contains any of the stable element in the matrix.

5.8.2.2 Alpha- and Beta-particle Measurement QC

Radiochemical blank samples (i.e., all components other than the original matrix material) are used to determine the amount of analyte contributed by the chemical reagents and physical processes used. Corrections of this amount of analyte radionuclide to the gross radionuclide determinations from the matrix samples are needed to determine the analyte radionuclide content from only the sample.

A radiochemical blank is run with each batch of samples. Batch sizes are generally three samples. The blank samples consist of all tracers, chemical yield monitors, and chemicals (except for the matrix material), and are subjected to the same radiochemical processing and measurement as the matrix samples.

5.8.2.3 Sample Dissolution and Radiochemical Separations

High temperature fusions or repeated treatments with aggressive acid mixtures (e.g., HF + HNO_3 + $HClO_4$) are used to achieve total dissolution and equilibration between the target radionuclides and their tracer [7-11].

Radiochemical separations are developed specifically for each natural-matrix SRM analysis [12-20]. The objective of the separation process is to produce counting sources of high radionuclide purity. To achieve this objective, a series of element specific chemical steps are employed that include co-precipitation, solvent extraction, chromatography, ion exchange, and redox techniques. The validation of the separation

procedure is evident by monitoring the isotopic purity of the resulting measurement spectrum, or confirmation of radioactive half-life.

5.8.2.4 Counting Source Preparation

Purified alpha-emitting element solutions are prepared for counting by either electrodeposition [21] or micro co-precipitation methods [22]. In general, electrodeposition of the alpha-emitting radionuclides is preferred. However, americium counting sources are best prepared by micro co-precipitation because of the added selectivity introduced by the fluoride precipitation step.

Purified ^{90}Sr is co-precipitated as strontium carbonate, filtered on a washed and pre-weighed low-ash high-retention 2.54-cm filter, washed with distilled water and 95% ethyl alcohol, vacuum dried, reweighed, mounted on 5-cm steel disc covered using double-stick tape, and covered with a Mylar film [23].

5.8.2.5 Instrumentation and Counting

Alpha-emitting sources for counting are measured under vacuum and in close geometry to silicon surface-barrier spectrometers [24]. Preset counting live times are selected to collect at least 10,000 counts per alpha peak, or two weeks, whichever occurs first. Energy calibrations of each alpha detector are determined by counting a variety of alpha-emitting radionuclides with well known alpha-particle emission energies. No detector efficiency determinations are needed because of the use of the isotope dilution tracer. Extended detector background measurements are made prior to and after measurement of samples.

Strontium-90 beta-particle emitting counting sources are measured using thin-window gas-flow proportional counters [25-26]. Each sample is counted continuously for approximately two weeks, with counting data stored in electronic files on a periodic basis (generally every 700 minutes) to follow the in-growth of the ^{90}Y daughter. Each detector is calibrated using a set of strontium carbonate standards containing ^{90}Sr in the same geometry as counting samples. The set of $SrCO_3$ standards are of varying mass to be used to estimate the appropriate counting efficiency for sample sources. Extended detector background measurements are made prior to and after measurement of samples.

5.8.2.6 Data Analysis: Peak Deconvolution Analysis, Interferences

Alpha spectra are analyzed for net peak area above the background using numeric methods that incorporate the assessment of peak shape and peak overlap [6]. Care is taken to account for radioactive impurities detected in the spectrum when determining the net peak areas.

The ^{90}Sr/^{90}Y in-growth curve is evaluated using the two component Bateman solution to the parent-daughter system [27-29].

5.8.2.7 Computations

Massic activity determinations from the alpha spectra are derived from the measurement model that includes contributions from the net peak counts, counting time, blank correction, activity of the tracer added to the sample, impurity corrections, emission probability, and radioactive decay corrections from the reference date [e.g., 30].

Massic activity determinations from the ^{90}Sr data are derived from the measurement model that includes contributions from the net counts, counting time, blank correction, chemical yield determined from the amount of stable Sr carrier initially added to the sample, impurity corrections, emission probability, counting efficiency calibrations, and radioactive decay corrections from the reference date [e.g., 23].

5.9 Interlaboratory comparison Data and Statistical Analysis

Resulting data from participating laboratories are carefully evaluated for systematic bias and material heterogeneity [31]. NIST accepts all data as valid unless there is strong scientific reasons that justify treating any identified datum as an outlier for rejection.

The protocol for data evaluation for each radionuclide is as follows:

DATA NORMALIZATION
Reference Date
Units
Expanded Uncertainty
DATA SCREENING
$\geq$ 3 Laboratories
Between-Laboratory Mean and Uncertainty (Mean Plot)
Between-Laboratory Data Distribution (Normal Probability Plot)
Distribution of Lab Means (Normal Probability Plot)
Resolve Interlaboratory Method Discrepancies
Laboratory Homogeneity (Normal Probability Plot)
DATA CERTIFICATION
Combined Data (Normal Probability Plot)
Bottle Number Heterogeneity (Curve Fitting)
Sample Size (*F*-Test)
Between-Bottle vs. Within Bottle Heterogeneity (*F*-test)
Distribution: Probability Plot Correlation Coefficient (PPCC)
Define Purposes for Material Use: Mean (e.g., Methods Development/validation), Tolerance (e.g., Proficiency Testing)
Bootstrap: Robustness Testing and data distribution characterization
Uncertainty Summary, including heterogeneity
Mean (k=2)
Tolerance Limit (95/95% confidence)

5.10 Certified and Uncertified Radionuclide Values

The protocol for evaluation of the interlaboratory comparison data had been carefully developed over the history of the SRM program [e.g., 32-33] in collaboration with the NIST Statistical Engineering Division. Acceptable data from a minimum of three laboratories are required to provide sufficient confidence for radionuclide massic activity certification. When there are statistically significant discrepancies among data sets, an uncertified mean value is provided, for information only, on the certificate.

The major evaluation issues that the radionuclide certification protocol focuses on include: a) systematic difference among analytical methodologies; b) consistency among the distributions of results from each laboratory; c) characterization of the distribution of the pooled results (including material heterogeneity and minor interlaboratory biases) by characterizing the mean value with its uncertainty; and d) the 95 percent tolerance limits. While normal probability plots allow identification of discrepant laboratory results, it is only through extensive personal contacts with the laboratory and additional revealing experiments that these discrepancies can be unfolded and resolved. Once the data are carefully screened, probability plot correlation coefficients are used to evaluate the robustness of characterizing the data with a number of distributional models and their family members. The criterion for the robustness of the data characterization is convergence of fit simulations, i.e., nearly identical results for the estimated mean and tolerance limit values, independent of the fit model chosen.

The strategies for certification of the massic activity of radionuclides in the natural-matrix material SRMs are summarized in 5.10.1.

5.10.1 Table of Natural Matrix Radionuclide SRM certification options:

Data Distribution	**Individual Data from Labs**	**Lab Means**	**Combined Data from Labs Grouped**	**Certify Mean**	**Certify uncertainty**	**Certify tolerance limit**
Normally Distributed	X	X		**X**	**X**	**X**
Normally Distributed	X	X	X	**X**	**X**	**X**
Normally Distributed	X			**Median of Lab Means**	**Median of Lab Means**	**X**
Non-Normally Distributed	X			**X**	**X**	**X**
Non-	X		X	**X**	**X**	**X**

Normally Distributed						

5.11 Certificate

The information (see attached Certificate) on the SRM certificate include,

- SRM number (inclusive of batch number) and name
- Description of the SRM and the intended and correct use of the material
- Unit mass
- Reference time
- Parties responsible for the preparation of material
- Source and preparation of the material
- Instructions for drying
- Radionuclide leachability
- Application of the certified values
- Uncertainties
- Heterogeneity determinations
- Notice and warning to users
  - Stability and expiration of certification (period of validity)
  - Radiological hazards
  - Storage and handling
- Contact persons
- Certified massic activities for radionuclides and uncertainties
  - Radionuclide
  - Mean + U (Bq g^{-1})
  - 2.5 to 97.5 percent tolerance limit (Bq g^{-1})
  - Number of assays
  - Half lives used
  - Methods
  - Contributing laboratories
- Notes
  - Analytical methods
  - Participating laboratories
- Uncertified massic activities
- Semi-quantitative trace element analysis
- Major elements recalculated as oxides

6 Uncertainty

The bootstrap method [34] is used to estimate the uncertainty for the natural-matrix SRM radionuclides certified massic activities. The bootstrap is a computationally-intensive, statistical procedure for estimating and computing the uncertainty of a statistic whose

form is complicated and/or whose underlying assumptions (e.g., normality) are non-standard. The bootstrap is utilized for the median calculations and for the tolerance limit calculations -- these statistics are distributionally complicated; also, the underlying normality of some of the data is suspect. The usual underlying assumptions do not hold due to a variety of experimental conditions, including interlaboratory biases, within-laboratory methodology differences, and material heterogeneity.

7 Equipment

Radionuclide measurement and balance equipment include:

- 5" NaI detectors (245/C17); “D” = 5-cm well, “E” = 2.5-cm well
- HPGe detectors, < 3 keV FWHM; (245/C13) = NIST # 519933; (245/C17) = PGT 1942 and 1945
- Alpha spectrometers (245/C13); <25 keV FWHM; NIST # 569445
- Gas-flow proportional counters (245/B001); <0.5 cpm background; NIST # 520379 and 619065
- Mettler 5-place balance (yearly Service and Calibration by Mettler using weights traceable to NIST); (245/C135) = NIST # 549813, (245/B152) = NIST # 594691
- Jupiter, 3-kg balance (yearly Service and Calibration by Mettler using weights traceable to NIST); (245/B152) = NIST # 526518

Calibrations of all radionuclide measurement equipment are discussed in section 5.8.1.1, 5.8.1.2, and 5.8.2.1.

8 Records

Data, analysis printouts, and copies of the SRM certificate are stored in folders identified by SRM number and held in the custody of the principle investigator.

Folders of SRM certificates are maintained in 245/C136 during the time of the SRM’s availability. When the SRM becomes unavailable, a copy of the SRM certificate remains in 245/C136.

Instrument calibration logs are maintained by the principle investigators in folders identified by SRM number.

Quality control logs are maintained by the principle investigators in folders identified by SRM number, and also entered into the instrument QC data book.

9 Filing and Retention

Data, analysis printouts and copies of the SRM certificate are stored in folders identified by SRM number and held in the custody of the principle investigator.

The IRD Quality Manager shall maintain the original and all past versions of this IRD Procedure.

Copies of the current revision of this Procedure shall be placed in controlled Quality Manuals. Electronic copies of this Procedure are uncontrolled versions.

All deleted Procedures (including old revisions) shall be maintained by the IRD Quality Manager. All old revisions shall be maintained until such time as it is decided to delete the Procedure. Once the decision has been made to delete the Procedure, only the last revision shall be maintained by the IRD Quality Manager.

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Appendix A: Natural Matrix Standard Needs

Matrix	Radionuclides	10-Yr Requirements
Soil: High Ca Low Ca	^{90}Sr, ^{137}Cs, ^{210}Pb, Alpha Emitters	5,000 Aliquants (1 kg Samples)
Sediments: High Ca Low Ca Mill Tailings	Alpha- & Beta-Particle, and Photon Emitters	1,600 Aliquants (100 g Samples)
Water: Aliquants	^{3}H, ^{60}Co, ^{90}Sr, ^{106}Ru, ^{134}Cs, ^{137}Cs, Natural Radionuclides, Alpha-Particle Emitters	Several Thousand (50-100 mL Samples)
Biological: Aliquants Lung Liver Bone Milk Sea Clam Sea Hare Seaweed	^{3}H, ^{14}C, Fission & Activation Products, Alpha-Particle Emitters	Several Hundred
Air: Filters	Natural Radionuclides	

Appendix B: Environmental Natural Matrix Standards

SRM No.	Name	Certified Massic Activity (Bq g^{-1})	Massic Activity given but not certified	Supple-mental Data
4350B	River Sediment	^{60}Co, ^{137}Cs, ^{152}Eu, ^{154}Eu, ^{226}Ra, ^{238}Pu, $^{239+240}$Pu, ^{241}Am	^{40}K, ^{55}Fe, ^{90}Sr, ^{228}Th, ^{230}Th, ^{232}Th, ^{234}U, ^{235}U, ^{238}U	a,c,d
4351	Human Lung	^{232}Th, ^{234}U, ^{238}U, $^{239+240}$Pu, ^{238}Pu/(^{239}Pu + ^{240}Pu)	^{228}Th, ^{230}Th, ^{241}Am	d
4352	Human Liver	^{238}Pu, ^{239}Pu + ^{240}Pu, ^{241}Am	^{228}Th, ^{230}Th, ^{232}Th, ^{234}U, ^{235}U, ^{238}U	d
4353	Rocky Flats Soil 1	^{40}K, ^{90}Sr, ^{137}Cs, ^{226}Ra, ^{228}Ac, ^{228}Th, ^{230}Th, ^{232}Th, ^{234}U, ^{238}U, ^{238}Pu, ^{239}Pu + ^{240}Pu, ^{241}Am	^{55}Fe, ^{235}U, ^{210}Pb	a,b,c,d
4354	Freshwater Lake	^{60}Co, ^{90}Sr, ^{137}Cs, ^{228}Th, ^{232}Th, ^{235}U, ^{238}U, ^{238}Pu, ^{239}Pu + ^{240}Pu, ^{241}Am	^{210}Pb, ^{226}Ra, ^{230}Th, ^{234}U	a,d
4355	Peruvian Soil	^{137}Cs, ^{228}Th, ^{230}Th, ^{232}Th, $^{239+240}$Pu, ^{241}Am, Upper limits on: ^{60}Co, ^{125}Sb, ^{152}Eu, ^{152}Eu, ^{154}Eu, ^{155}Eu	^{40}K, ^{55}Fe, ^{90}Sr, ^{208}Tl, ^{214}Bi, ^{238}Pu	d
4356	Ashed Bone			
4357	Ocean Sediment	^{40}K, ^{90}Sr, ^{137}Cs, ^{226}Ra, ^{228}Ra, ^{228}Th ^{230}Th, ^{232}Th, ^{238}Pu, ^{239}Pu + ^{240}Pu	^{129}I, ^{155}Eu, ^{210}Po, ^{210}Pb, ^{212}Pb, ^{214}Bi, ^{234}U, ^{235}U, ^{237}Np, ^{238}U, ^{241}Am	a, d

a) Semi-quantitative elemental analysis by emission spectrographic measurements.
b) Semi-quantitative analysis of mineral components by x-ray diffraction measurements.
c) Analysis of plutonium isotopes by mass spectrometry.
d) Particle size distribution.

Appendix C: Approval Form for SRMs

TECHNICAL REVIEW - SRM CERTIFICATES

REVIEW THE ATTACHED DRAFT CAREFULLY. ANY SUGGESTIONS OR CHANGES SHOULD BE MADE ON THIS DRAFT. FORWARD DRAFT AND APPROVAL RECORD TO THE NEXT INDIVIDUAL INDICATED.

RETURN TO: Standard Reference Materials Program, Stop 2321

SRM: 4334H

TITLE: Plutonium-242 Solution Radioactivity Standard

DATE: 04 January 2005

PROJECT MANAGER: _____

TELEPHONE: _____

ROUTING				APPROVAL	
NAME (REVIEWER)	BLDG. ROOM	DIV.	SEC.	INITIALS	DATE
K. Inn	245 C114	846	04	KGWI	18 Jan'05
M.P. Unterweger	245 C114	846	04		
L.R. Karam	245 C235	846	00		
L.L. Lucas	245 C114	846	04		

Appendix D:

National Institute of Standards and Technology

Certificate

Environmental Radioactivity

Standard Reference Material 4357

Ocean Sediment Radionuclide Standard

Description of the Standard and Intended Use: This Standard Reference Material (SRM) has been developed in cooperation with member laboratories of the International Committee for Radionuclide Metrology and expert national laboratories. The SRM is intended for use in tests of measurements of environmental radioactivity contained in matrices similar to field samples. Uses of the material include: 1) development of radiochemical procedures, 2) test of radiochemical procedures already in use for environmental and biokinetic evaluations, 3) calibration of instruments, 4) interlaboratory comparison materials for radiochemical methods evaluation, 5) test for competency of technicians to do radiochemical assays, and 6) demonstration that data output is reliable.

Unit Mass: 85 g Nominal
Reference Time: 16 February 1994

Preparation of Material: This Standard Reference Material was prepared in the Physics Laboratory, Ionizing Radiation Division, Radioactivity Group, J.M.R. Hutchinson, Group Leader. The overall technical direction leading to certification was provided by Kenneth G.W. Inn of the Radioactivity Group.

Statistical support was provided by Drs. James J. Filliben, Eric S. Lagergren, Walter S. Liggett, Nien-Fan Zhang, and Keith R. Eberhardt.

The support aspects involved in the preparation, certification, and issuance of this SRM were coordinated through the Standard Reference Materials Program by Nancy M. Trahey.

Gaithersburg, Maryland 20899
January 1996

Thomas E. Gills, Chief
Standard Reference Materials Program

Source and Preparation of Material: The sediment is a blend of material collected off the coast of Sellafield, UK, and in the Chesapeake Bay, USA, in the weight ratio of 1:200, respectively. A. Knight and M. Measures of the National Radiological Protection Board, UK, collected, sieved to -200 mesh, dried and analyzed the Sellafield Sediment before sending it to NIST. The Chesapeake Bay sediment was freeze dried, blended with the Sellafield sediment, sterilized with 50 kGrays of ^{60}Co radiation and pulverized with a "pancake"-style air-jet mill. The average particle diameter for the resulting powder is approximately 6 μm, and more than 99 percent, by weight, of the particles are less than 20 μm in diameter.

Instructions for Drying: When nonvolatile radionuclides are to be determined, working samples of this SRM should be dried in air at 40°C for 24 hours prior to weighing. Volatile radionuclides (e.g., ^{137}Cs, ^{210}Pb and ^{212}Pb) should be determined on samples as received; separate samples should be dried as previously described to obtain a correction factor for moisture. Correction for moisture content is to be made to the data for volatile radionuclides before comparing to the certified values. This procedure ensures that these radionuclides are not lost during drying. The weight loss on drying is typically less than 2 percent.

Radionuclide Leachability: All actinides and their daughters are approximately 87 percent removed from the sample by normal HNO_3 or HNO_3-HCl leaching procedures. Total sample digestion or non-destructive analysis is required to produce results that can be compared to those listed in this certificate.

Application of the Certified Values: When 5 or more measurements are available, compute the sample mean and ascertain that the mean falls within the certified mean plus uncertainties interval. When 4 or fewer measurements are available, then ascertain that all of the individual values are within the certified tolerance limits interval.

Uncertainties: The bootstrap is a computationally-intensive, statistical procedure for estimating and computing the uncertainty of a statistic whose form is complicated and/or whose underlying assumptions (e.g., normality) are non-standard. The virtue of the procedure is that it provides a straightforward, rigorous methodology for computing uncertainties that would otherwise have been difficult to obtain.

The bootstrap was utilized here for the median calculations and for the tolerance limit calculations -- these statistics are distributionally complicated; also, the underlying normality of some of the data is suspect. The usual underlying assumptions do not hold due to a variety of experimental conditions, including interlaboratory biases, within-laboratory methodology differences, and material heterogeneity.

Procedurally, the bootstrap estimate for the uncertainty of a statistic (e.g., the median) is obtained as follows:

1. From the original sample of n observations, compute the statistic of interest (e.g., the median).

2. From the original n data points, extract a random sample -- with replacement -- of n points (this becomes the "bootstrap sample").

3. Compute the statistic of interest (e.g., the median) from this bootstrap sample (this will be the bootstrap statistic).

4. Repeat steps 2 and 3 a large number of times (e.g., 1000 times); the bootstrap statistic will, of course, change from one bootstrap sample to the next.

5. Compute the standard deviation of the statistic by applying the usual standard deviation formula to the 1000 bootstrap statistics.

Reference: Efron B. and Tibshirani, R.J. (1993). An Introduction to the Bootstrap. Monographs on Statistics & Applied Probability 57, Chapman and Hall, New York.

Heterogeneity Determinations: The material has been tested for sample sizes of 10 to 100 grams, for which the heterogeneity of gamma-ray-emitting radionuclides have been detectable. Furthermore, material heterogeneity has been detected at a sample size of 10 grams for ^{90}Sr and actinide radionuclides. The expected variation of measurements due to heterogeneity have been incorporated in the certified tolerance limits and uncertainty of mean concentration values. The certified values for radionuclides with normal distribution of analytical measurements are listed in Table 1. Table 2 lists the certified values for radionuclides with non-normal distribution of analytical measurements. It is recommended that a sample sizes of 10 grams or larger be used for radiometric and radiochemical analysis.

Notice and Warnings to Users:

Stability and Expiration of Certification: This matrix is considered to be stable; however, its stability has not been rigorously assessed. NIST will monitor this material and will report any substantive changes in certification to the purchaser. Should any of the certified values change, purchasers will be notified by NIST. Return of the attached registration card is mandatory to receive such notifications.

Radiological Hazards: The SRM contains low levels of anthropogenic and natural radioactivity. The SRM poses no radiological hazard. The SRM should be used only by qualified quality control persons.

Chemical Hazards: The SRM is a dried sterilized sediment and poses no chemical hazard. However, inhalation or ingestion of the material is not recommended.

Storage and Handling: The SRM should be stored in a dry location at room temperature. The bottle should be shaken before opening in a chemical hood, and the bottle should be recapped tightly as soon as subsamples are removed. The SRM should always be clearly marked and should be packed, marked, labeled, and shipped in accordance with the applicable national, international, and carrier regulations if it needs to be transported.

Contact Persons: For further information contact Zhichao Lin (zclin@micf.nist.gov; phone: 1-301-975-5645) or Kenneth G.W. Inn (e-mail: kenneth.inn@nist.gov; phone: 1-301-975-5541), NIST, Building 245, Room C114, Gaithersburg, MD 20899, fax 1-301-869-7682

Table 1: Certified Massic Activities for Radionuclides with Normal Distribution of Results

Radio-nuclide	Mean $\pm$ $2s_m$* ($mBq \cdot g^{-1}$)	2.5 to 97.5 Percent Tolerance Limit** ($mBq \cdot g^{-1}$)	Number of Assays	Half Lives Used (a)***	Methods	**Contributing Laboratories**
^{40}K	225 $\pm$ 5	190 - 259	31	1.277×10^9 $\pm$ 8×10^6	3a	KU, MAFF, NIR, NPL, YAEL
^{226}Ra	12.7 $\pm$ 0.4	10.3 - 15.0	21	1600 $\pm$ 7	3a	EML, IT, KU, NPL
^{228}Ra	13.3 $\pm$ 0.8	9.2 - 17.4	20	5.75 $\pm$ 0.03	3a	IT, KU, NPL
^{228}Th	12.1 $\pm$ 0.3	9.7 - 14.6	40	1.9131 $\pm$ 0.0009	3a +1c	EML, KU, LGC, MAFF, NIST, NPL, OSUH, YAEL
^{230}Th	12.0 $\pm$ 0.5	9.6 - 14.4	18	75380 $\pm$ 300	1c	LGC, NIST, OSUH
^{232}Th	13.0 $\pm$ 0.3	11.6 - 14.3	18	1.405×10^{10} $\pm$ 6×10^7	1c	LGC, NIST, OSUH

* Two standard deviations of the Mean

** Normal Tolerance Limit for 95 percent confidence and 95 percent coverage.

*** Evaluated Nuclear Data Structure File (ENSDF), January 1996. The stated uncertainty is the standard uncertainty.

Table 2: Certified Massic Activities for Radionuclides with Non-Normal Distribution of Results

Radio-nuclide	Mean ± $2s_m$* ($mBq \cdot g^{-1}$)	2.5 to 97.5 Percent Tolerance Limit** ($mBq \cdot g^{-1}$)	Number of Assays	Half Lives Used (a)***	Methods	Contributing Laboratories
^{90}Sr	4.4 ± 0.3	2.1 - 8.4	49	28.78 ± 0.04	1b, 2d	EML, LGC, MAFF, NE, NRPB, YAEL
^{137}Cs	12.7 ± 0.2	10.8 - 15.9	76	30.07 ± 0.03	3a	EML, IT, KU, LGC, MAFF, NE, NIR, NPL, ORNL, OSUB, YAEL
^{238}Pu	2.32 ± 0.06	2.01 - 3.02	53	87.7 ± 0.3	1c	EML, LGC, MAFF, NIST, OSUB
^{239}Pu + ^{240}Pu	10.4 ± 0.2	9.2 - 13.3	72	24110 ± 30 6564 ± 11	3a +1c	EML, IT, LGC, MAFF, NIST, OSUB, OSUH

* Two standard deviations about the Weibull Mean

** Weibull Tolerance Limit for 95 percent confidence and 95 percent coverage

*** Evaluated Nuclear Data Structure File (ENSDF), January 1996. The stated uncertainty is the standard uncertainty.

NOTES:

Analytical Methods:

1. HF-HNO_3 or HF-HNO_3-$HClO_4$ Dissolution
2. NaOH-HCl Leach
3. Non-Destructive Analysis

a. Germanium Gamma-ray Spectrometer
b. Thin-Window Beta-Particle Geiger Counter
c. Silicon Surface-Barrier Alpha-Particle Spectrometer
d. Plastic-Phosphor Beta-Particle Scintillation Counter

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South of Scotland Electricity Board (SSEB)
Peter Young
Hunterston, UK

Yankee Atomic Environmental Laboratory (YAEL)
David McCurdy
Bolton, MA, USA

[5]Retired

[6]Now at USDOE Environmental Measurements Laboratory, NY

Uncertified Massic Activities: Radionuclides for which insufficient numbers of data sets, or for which discrepant data sets were obtained, are listed in Table 3. No uncertainties are provided because insufficient bases upon which meaningful estimates could be determined.

Table 3: Uncertified Massic Activities

Radionuclide	Massic Activity ($mBq \cdot g^{-1}$)	Methods	Contributing Laboratories
^{129}I	0.009	3a +1c	NIR
^{155}Eu	1.4	3a	MAFF
^{210}Po	14	3a	OSUH
^{210}Pb	24	3a	IT, ORNL
^{212}Pb	14	3a	MAFF
^{214}Bi	14.5	3a	MAFF
^{234}U	12	3a +1c	AWE, IT, LGC, OSUH, NIST
^{235}U	0.40	3a +1c	AWE, LGC, NIST, NPL
^{238}U	12	3a +1c	IT, LGC, NIST, OSUH
^{237}Np	0.007	3a +1c	LRM, KU
^{241}Am	10	3a +1c	AWE, BNF, IT, KU, LGC, MAFF, NIST, NPL, ORNL, OSUB

Semi-Quantitative Trace Element Analysis:

Tables 4 and 5 are intended for information only. The values given are not certified.

Table 4: Semi-quantitative Emission Spectrographic Analysis for SRM 4357

Element	**µg ·g^{-1}**	Element	µg ·g^{-1}	Element	µg ·g^{-1}
Ag	0.12	Al	24700	As	<100
Au	<6.8	B	34	Ba	143
Be	<0.1	Bi	<10	Ca	6267
Cd	<32	Ce	<43	Co	2.9
Cr	27	Cu	82	Dy	<22
Er	<4.6	Eu	<2.2	Fe	10700
Ga	3.5	Gd	<32	Ge	<4.6
Hf	<150	Ho	<6.8	In	<10
Ir	<15	K	5070	La	25
Li	<68	Lu	<15	Mg	3930
Mn	163	Mo	1.8	Na	4000
Nb	10	Nd	<32	Ni	97
Os	<15	P	<680	Pb	12
Pd	<1.0	Pr	<100	Pt	<2.2
Re	<10	Rh	<2.2	Ru	<2.2
Sb	<68	Sc	2.8	Si	>340000
Sm	<10	Sn	<4.6	Sr	64
Ta	<320	Tb	<32	Th	<46
Tl	<10	Tm	<4.6	U	<220
V	21	W	<15	Y	12
Yb	1.8	Zn	45	Zr	540

Table 5: Major Elements Recalculated as Oxides

Oxide	Weight Percent	Oxide	Weight Percent
SiO_2	>73	Al_2O_3	4.7
Fe_2O_3	1.5	MgO	0.65
CaO	0.88	Na_2O	0.54
K_2O	0.5	TiO_2	0.42
P_2O_5	<0.16	MnO	0.021